

**CONFIDENTIAL INFORMATION
SUBJECT TO PROTECTIVE ORDER**

**IN THE UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA**

ELF ATOCHEM NORTH AMERICA, INC.)
Plaintiff,)
v.) Civil Action No. 1:99CV02559(TPJ)
Q. TODD DICKINSON,)
Commissioner of Patents)
and Trademarks)
Defendant.)

**DECLARATION OF DR. CHRISTOPHER A. BERTELO
SUBMITTED PURSUANT TO FED. R. CIV. P. 56(a) AND LOCAL RULE 11.2
IN SUPPORT OF PLAINTIFF'S OPPOSITION TO DEFENDANT'S MOTION FOR
SUMMARY JUDGMENT**

I, Christopher A. Bertelo, do hereby declare that:

1. I obtained a Ph.D. in chemistry in 1975 from Princeton University. After receiving my doctorate, I took a position as an Associate Research Chemist at the University of Iowa for two years. In 1977, I taught chemistry as a visiting assistant professor at Grinnell College in Iowa. From 1978-1985, I worked as a senior chemist for Tenneco Chemical Co. in basic research, polyvinyl chloride polymer process development, and plastics additives research.
2. Since 1985, I have worked for Elf Atochem North America, Inc. ("Atochem"), formerly M&T Chemicals, Inc. I am currently the Manager of Research and Development for the Plastic Additives Business of Atochem. I have held this position for 5 years. Prior to my current position,

I was the project leader for Atochem's Impact Modifier Research and Development Program. I have worked directly in stabilizer research and in polymer research in these positions and in other business at Atochem.

3. In total, I have over 20 years experience in industrial chemistry. My curriculum vitae is attached as Ex. U.

I. The Chenard Application and the Prior Art

4. I understand that Atochem filed this lawsuit against the U.S. Patent & Trademark Office (the "PTO") after it rejected a patent application of Dr. Jean-Yves Chenard and Dr. Jean-Claude Mendelsohn. [See U.S. Patent App. No. 07/870,759 ('759 application), Ex. D.]

5. I have reviewed the '759 application and I am aware that it discloses a mixture of a chemical compound of the type called "mercapto alkanol esters" (also called "reverse esters") and certain organotin compounds (collectively, the "claimed invention") and the mixture's use for stabilizing plastics of the class known as polyvinyl halide ("PVH") polymers, which includes the widely used plastic material PVC (polyvinyl chloride, one type of PVH polymer).

6. The mercapto *alkanol* esters are called "reverse esters" to distinguish them from mercapto *acid* esters. The organotin compounds that are included in the claimed compositions are organometallic compounds which contain tin bonded to both a carbon and a sulfur atom (the claimed organotin-sulfur compounds) or tin bonded to both a carbon and a halogen (typically chlorine or fluorine) atom (the claimed organotin-halide compounds).

7. It is my understanding that the PTO has rejected the claimed invention as "obvious" over the prior art, particularly U.S. Patent No. 3,928,285 by Gough et al. (the "Gough patent"). I am familiar with the Gough patent; it is attached as Ex. F. I understand that the PTO alleges that each component of the claimed invention is known in separate prior art references and that it would have been obvious to one of skill in the art to select the known components from these references and combine them to make the claimed combinations.

8. Specifically, I understand that the PTO has alleged that the broad general disclosure of thiol compounds in the Gough patent, which includes reverse esters, would have motivated someone of ordinary skill in the art to select this one small class of compounds out of the infinite number of compounds within the many classes of thiols. The PTO then alleges that one of skill in the art would have been motivated by other references to replace the disclosed organotin-borates of Gough with the organotin-sulfur or organotin-halide stabilizers.

9. In my opinion, PVH stabilization researchers at the time of the Chenard and Mendelsohn invention in the late 1970s would have understood that Gough discloses the use of a very specialized class of tin compounds called organotin-borates. Gough generally teaches that these organotin-borates may be improved as stabilizers by the addition of any thiol (SH) compound. Gough does not disclose, however, that reverse esters would work any better than the other classes of thiols disclosed in Gough, let alone the claimed reverse esters.

10. In addition, I am familiar with the declaration by Dr. Michel Foure in 1982 [Ex. P], which was submitted to the PTO in support of the patentability of the claimed invention. I am familiar with the testing procedures described in the 1982 Foure Declaration.

II. Experiments Performed in Support of Patentability

11. I supervised several recent experiments in support of the claimed invention. I used the same procedures and amounts of reagents used by Dr. Foure in the 1982 declaration. The exact conditions used for the tests reported in the 1982 Declaration cannot be repeated today. As a general matter for all laboratory experiments, different conditions in a laboratory on any one day (such as atmospheric conditions or reagent quality) can influence the results for sets of experiments performed on that day. Nevertheless, the results of my experiments are consistent with and will illustrate the same general trends as those seen in Dr. Foure's experiments. The trends I observed confirm and expand upon those observed by Dr. Foure.

12. My tests provide additional comparisons to those in the 1982 Declaration. Specifically, I tested combinations of an organotin-borate compound (from the Gough patent) with the claimed reverse esters, as well as with a mercapto acid ester used in Gough. The organotin-borate compound is one in which a tin atom is bonded to an oxygen atom, which is then bonded to a boron atom.

13. Gough did not test any combinations of organotin-borates with any reverse esters; he only tested combinations of organotin-borates with simple thiols and mercapto acid esters.

14. The 1982 Declaration indicates that Dr. Foure used a device called a "colorimeter" which objectively measures differences in color or shading in PVH plastic and assigns different numerical values to samples on the basis of the degree and type of discoloration of those samples. The coloring or shading of a sample of PVH plastic, such as PVC, is an indication of the stability of that plastic; discoloration reflects chemical degradation from the heat and stress of manufacturing.

Because PVC is susceptible to degradation during processing, stabilizers are included to prevent degradation. The colorimeter is a commonly used analytical device which measures the stability achieved by different stabilizers. In the testing reported in his Declaration, Dr. Foure used a particular device identified as the Macbeth MC 1500 S colorimeter, which gave "whiteness index values." These values are simply a quantitative measure of how "white" a particular PVH sample is. The Macbeth MC 1500 S has been replaced in Atochem's laboratory with a newer-model colorimeter called a ColorQuest II Model C 5405. The colorimeter results are reproducible to less than ± 0.1 unit. A trained observer can visually distinguish 1 unit variations in whiteness index values.

15. In addition to whiteness index values, the ColorQuest II Model C 5405 colorimeter provides yellowness index values, still another measure of discoloration. In general, the whiteness index is a measure of how white (desirable) something is, and the yellowness index is a measure of how yellow (undesirable) something is.

16. Both whiteness and yellowness values are calculated from L a* b* values, which represent independent (non-overlapping) color measurements. Those in the PVH stabilization industry, for example, know that the L value represents the intensity of light reflected from the sample (0 = no reflection, 100 = total reflection); the a* value represents the red-green spectrum (whether and to what extent the sample has red and green in it); and the b* value represents the blue-yellow spectrum (whether and to what extent the sample has blue and yellow in it). An explanatory diagram of the color measurements is attached as Ex. V.

17. The whiteness and yellowness index values are each calculated from the L a*b* values, as shown in Ex. W. Thus, for a given set of L a* b* values, researchers can calculate both a whiteness index and a yellowness index value.

18. The L a* b* values for my experiments are attached as Ex. Q.

19. The L a* b* values are considered by those in the PVH industry as a standard measure of polymer degradation. In general, as the whiteness value increases, the yellowness value decreases.¹

20. I agree with Dr. Foure's statement in his 1982 Declaration that a difference of 3 whiteness index units is important and a difference of 5 or more units is significant. By "important," I mean that those in the PVH industry would be able to visually distinguish the different stabilization effects and would consider the higher whiteness-value stabilizer to impart better stabilization. The higher whiteness-value stabilizer would also be recognized for its commercial potential to provide acceptable stabilization at a reduced tin content. By "significant," I mean that those in the PVH industry would be even more compelled by the commercial potential of the clearly superior, higher whiteness-value stabilizer.

21. I also consider a difference of 3 yellowness index values important and a difference of 5 yellowness index values significant for the same reasons as described above. A difference of 3 yellowness index units for different stabilizer compositions is visually distinguishable and commercially important. A stabilizer with a yellowness index value 5 units lower than another stabilizer has stabilized a PVH polymer significantly better and presents a substantial opportunity

¹ In some circumstances, however, especially for high a* values and low L values, the yellowness index can decrease while the whiteness index also decreases. This phenomenon occurs because the different equations for whiteness and yellowness index values assign different weight to the L a* b* values.

to reduce the overall tin content of the stabilizer while still attaining acceptable levels of stabilization.

22. Colorimeters such as the ColorQuest II Model C 5405 give better results for whiteness index ranges above about 30. Although the colorimeter calculates values for samples below zero, such samples are so badly degraded that they are of little experimental or commercial interest. Further, the calculated distinctions between low values are not as reliable on an absolute scale, but low values are a reliable indicator of a badly degraded polymer. I would thus consider differences of 3 units to be especially important and differences of 5 units to be especially significant between samples with high whiteness index values.

23. Twelve experiments were performed:

a. I determined the stabilization performance of combinations of an organotin-borate (from the Gough patent and not within the scope of the claimed invention) with each of three different sulfur compounds. The organotin-borate tested was the product (a-1) of the Gough patent [Ex. F] used in Examples 20 to 27 (in columns 13-14, Table III). The three sulfur compounds are 2-mercaptopropyl stearate ("MES", within the scope of the claims), 2-mercaptopropyl octanoate ("MEO", within the scope of the claims), and octadecyl mercaptoacetate ("ODMA", outside the scope of the claims but used in the Gough patent in Example 22, columns 13-14, Table III). The results for my experiments are reported in Tables 1-4 and Figures 1-4 below.

b. I also determined the stabilization performance of combinations of two other organotin-sulfur compounds designated "T₁" and "T₂" within the scope of the claimed invention with each of the same three sulfur compounds referred to in subparagraph (a) above.

c. I selected two of the sulfur compounds to demonstrate that the only possible variable that could account for any difference in performance would be the position of the SH group relative to the (CO₂) ester group in the molecule, a distinguishing characteristic of the claimed reverse esters. These results are reported below in Tables 1-4 and Figures 1-4.

d. I tested the chemicals as follows:

- i. Gough organotin-borate alone (outside the scope of the claims)
- ii. Gough organotin-borate with each of 3 sulfur compounds
- iii. T₁ organotin-sulfur alone (within the scope of the claims)
- iv. T₁ organotin-sulfur with each of 3 sulfur compounds
- v. T₂ organotin-sulfur alone (within the scope of the claims)
- vi. T₂ organotin-sulfur with each of 3 sulfur compounds

e. One of these claimed compounds (T₁) had nearly the same amount of tin (about 29%) as the Gough organotin-borate (about 31%).² This comparison between tin compounds with nearly equivalent percentages is especially meaningful because tin level is the most important indicator of probable stabilization performance to those in the PVH stabilization industry. The results for these control experiments without sulfur compounds are reported in Tables 5-6 and Figures 5-6 below.

f. I also compared the stabilization performance of the Gough organotin-borate with a second claimed tin compound, T₂, which had about half the tin content (about 14.5%) as the organotin-borate (31%). These experiments were designed to demonstrate the efficacy of the

² These tin percentages are calculated by dividing the molecular weight of tin (about 119 for each atom) in the molecule and dividing it by the molecular weight of the entire tin stabilizer molecule.

claimed invention even with a significant reduction in tin content. The results are of extreme commercial importance because tin is generally the most expensive component of stabilizer compositions. The ability to reduce tin content and still give effective stabilization translates into significant cost savings. The results for these tin compounds in combination with sulfur compounds are reported in Tables 3-4 and Figures 3-4 below.

24. The three sulfur compounds that I tested are set forth with molecular formulas below:

(1) 2-mercaptopethyl stearate $C_{17}H_{35}CO_2CH_2CH_2SH$

(a reverse ester within the scope of the claimed invention, abbreviated as MES)

(2) 2-mercaptopethyl octanoate $CH_3(CH_2)_6CO_2CH_2CH_2SH$

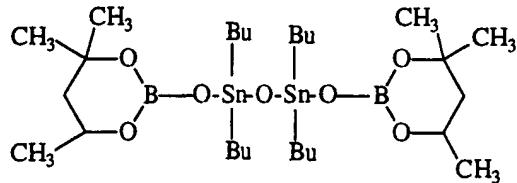
(a reverse ester within the scope of the claimed invention and listed in Gough's specification at column 8, line 64, but not tested by Gough, also called 2-thioethyl octanoate and abbreviated as MEO)

(3) octadecyl mercaptoacetate $HSCH_2CO_2C_{18}H_{37}$

(a mercapto acid ester not within the scope of the claimed invention, abbreviated as ODMA, also called stearyl mercaptoacetate or octadecylthioglycolate)

25. I tested each of the MES, MEO and ODMA sulfur compounds in respective combinations with the following tin compounds:

(1) Gough organotin-borate compound (outside the scope of the claimed invention):

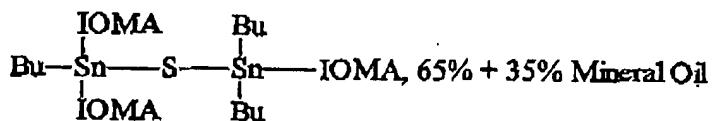


This is an organotin-borate, the product a-1 (Example 1) in Gough, is used in Examples 20 to 27 of Gough (columns 13 and 14, Table III). It has a tin content of about 31%.³

(2) $T_1: \text{BuSn}(\text{S})\text{SC}_{12}\text{H}_{25}$ monobutyltin (sulfide)(dodecylmercaptide)

This organotin-sulfur compound is within the scope of the claimed invention. It has a tin content of about 29%.

(3) $T_2:$



IOMA = isoocetylmercaptoacetate

This organotin-sulfur compound is within the scope of the claimed invention. It has a tin content of about 14.5%.

³ Gough called this compound 1, 3-bis(4, 4, 6-trimethyl-1, 3, 2-dioxaborinyl-2-oxy)(tetra-n-butyltinannoxane) ("a-1"). He provided a structure for this compound in column 6, lines 40-50. Although I do not believe that Gough's nomenclature accurately describes the structure of the (a-1) compound, I assume that the structure provided is in fact the compound (a-1) which was used and tested in Gough.

III. Experimental Results and Conclusions

26. For the tests reported in Tables 1-4 and Figures 1-4, I combined 0.15 phr (parts per hundred parts PVH polymer resin) of the tin compound with 0.3 phr sulfur compound. These were combined with a master batch of PVC according to the protocol set forth in the 1982 Declaration by Dr. Foure.

27. I compared the stabilization results for (1) unclaimed combinations of an organotin-borate (from Gough) and reverse ester sulfur compounds with (2) claimed combinations of an organotin-sulfur compound (T_1) and reverse esters. The claimed organotin-sulfur compound has a tin content which is approximately equivalent to the tin content of Gough's organotin-borate compound (which is outside the scope of the claims). I also tested these tin compounds with a sulfur compound outside of the claimed invention, ODMA. These results are shown below in Table 1.

TABLE 1
Whiteness Index Values for Stabilizer Combinations: T_1 and Organotin-borate

| Sulfur Compounds | T_1 + Sulfur Compound | Organotin-borate + Sulfur Compound |
|----------------------------------|-------------------------|------------------------------------|
| 2-mercaptoethyl stearate (MES) | 40.3* | -13.7 |
| 2-mercaptoethyl octanoate (MEO) | 45.7 | -9.5 |
| octadecyl mercaptoacetate (ODMA) | 9.2 | -22.6 |

Note: The *higher* the number, the better the result.

* Results in bold pertain to claimed combinations.

28. Table 1 shows that all combinations of an organotin-borate with the sulfur compounds give terrible results. Negative values are unacceptable to those in the polymer industry. The results for the claimed combinations (shown in bold-face type), however, are significantly superior to the

results for combinations of organotin-borates. The results are especially surprising because T_1 and the organotin-borate have approximately the same tin content, about 30%. For commercial stabilizers, tin content is the best indicator both of stabilization performance and of cost. Clearly, the claimed combinations (whiteness index values of 40.3 and 45.7) are superior to combinations of organotin-borates with the same sulfur compounds, which give negative whiteness index values of -13.7 and -9.5.

29. Table 1 also reports that the combinations of the claimed invention are significantly superior to combinations of the organotin-sulfur compound (T_1) with a sulfur compound outside the scope of the claimed invention, ODMA. The values for the claimed reverse esters (45.7 and 40.3) are significantly higher than that for ODMA (9.2).

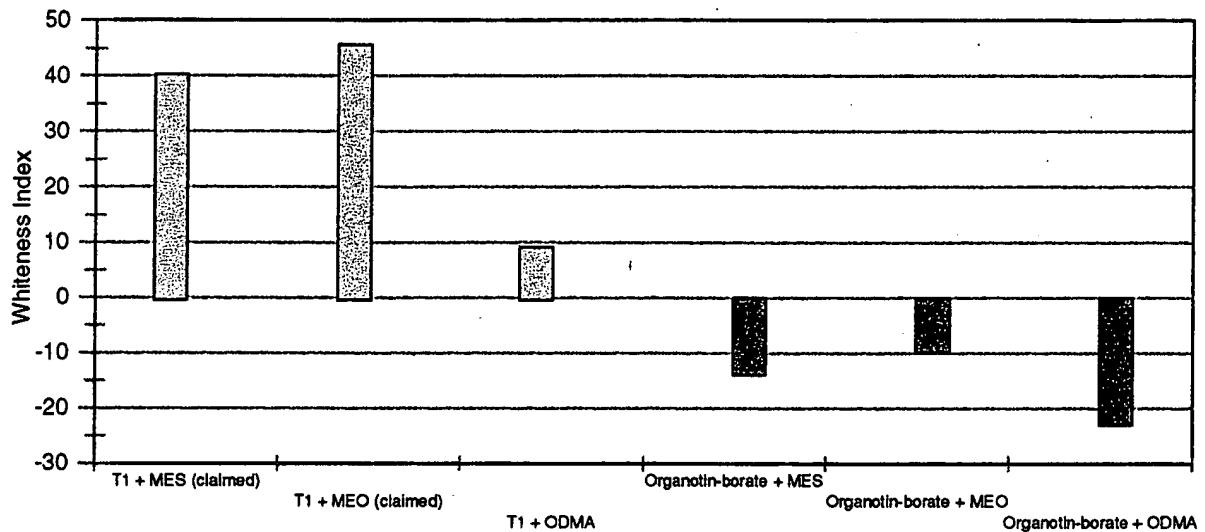
30. In my opinion, the disparate results for these two sulfur compounds can only be explained by the position of the SH group in the molecule relative to the ester (CO_2) group.

MES $C_{17}H_{35}CO_2CH_2CH_2SH$ W.I. = 40.3

ODMA $HSCH_2CO_2C_{18}H_{37}$ W.I. = 9.2

31. As the molecular formulas of these compounds show, the *only significant* structural difference between these two sulfur compounds is the position of the SH group in the compound. Each compound has 20 carbon atoms and a very similar arrangement of atoms. In my opinion, it would have been completely unexpected that this mere positional difference of SH in the molecule would yield a difference in stabilization of (40.3 - 9.2) **31.1** total units.

FIGURE 1
Whiteness Index Values for Stabilizer Combinations: T₁ and Organotin-borate



32. Figure 1 visually demonstrates the significance of the values in Table 1. The two claimed combinations on the far left side of Figure 1 are significantly better than all combinations outside of the claimed invention, especially the combination of an organotin-borate compound with the same three sulfur compounds.

33. Gough did not test the organotin-borate compounds in combination with reverse esters of the claimed invention. Based on the test results I obtained with the organotin-borate, it is my opinion that if such tests had been performed by Gough or by someone in the industry at that time, the results would not have suggested to anyone that reverse esters were superior to other thiol compounds in Gough. Although the values for an organotin-borate with the claimed MES and MEO sulfur compounds are a little better than values for the organotin-borate with ODMA (a sulfur

compound not within the scope of the claimed invention), the results are still unacceptable from both an experimental and a commercial standpoint. Nothing in Gough points to the superiority of the reverse esters that Chenard and Mendelsohn discovered. Rather, the reverse esters would have been dismissed as unacceptable along with the other sulfur compounds when used in combination with Gough's organotin-borates. In other words, there is no indication in Gough that distinguishes the claimed reverse esters over other thiols.

34. The claimed reverse esters did not show signs of superiority until the inventors tested them with organotin-sulfur and organotin-halogen compounds in the claimed combinations. This testing dramatically illustrated the superiority of the reverse esters when used in the claimed combinations.

35. Table 2 presents yellowness index results for the same combinations as Table 1.

TABLE 2
Yellowness Index Values for Stabilizer Combinations: T_1 and Organotin-borate

| Sulfur Compounds | T_1 + Sulfur Compound | Organotin-borate + Sulfur Compound |
|----------------------------------|-------------------------|------------------------------------|
| 2-mercaptoethyl stearate (MES) | 15.3 | 34.7 |
| 2-mercaptoethyl octanoate (MEO) | 13.4 | 32.6 |
| octadecyl mercaptoacetate (ODMA) | 26.1 | 39.1 |

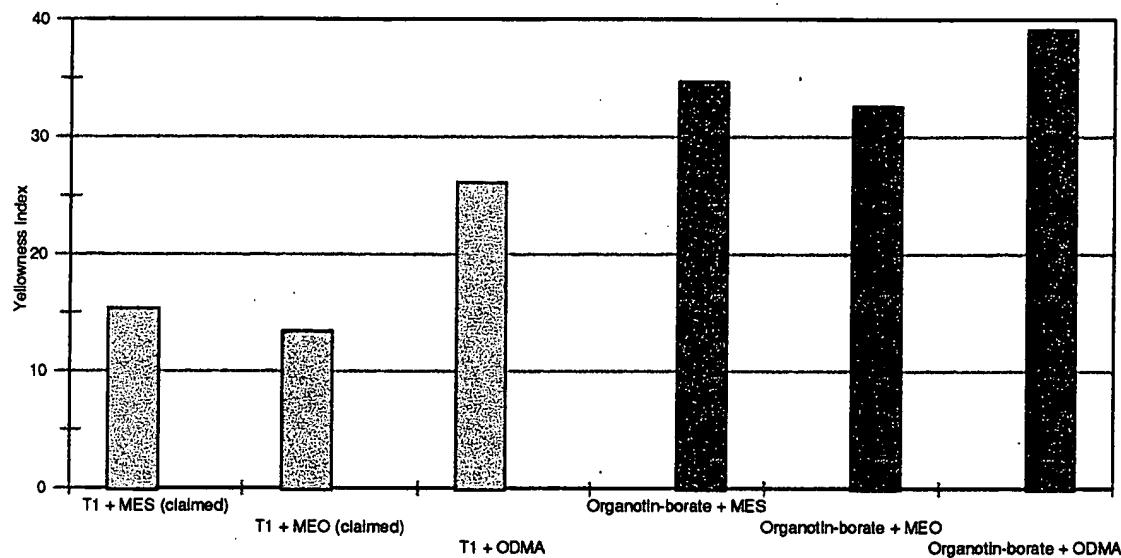
Note: The *lower* the number, the better the result.

36. The yellowness index values in Table 2 confirm the whiteness index results of Table 1 for the same combinations of compounds. Results for the claimed combinations, shown in bold-face type, are significantly superior to the combinations of organotin-borates. Despite having nearly equivalent tin content (about 30%), the claimed combinations with yellowness index values of 15.3

and 13.4 are superior to combinations of organotin-borates with the same sulfur compounds, which yield higher (less desirable) yellowness index values of 34.7 and 32.6.

37. Combinations of the claimed invention are also superior to a combination of T_1 with a sulfur compound that is not within the scope of the invention, ODMA. The above yellowness index values show that the claimed reverse ester combinations allow significantly less discoloration than the combination of T_1 with ODMA. In my opinion, nothing in the Gough patent taught that reverse esters would give superior stabilization performance, relative to Gough's other thiol compounds, in stabilizer compositions with the claimed organotin compounds.

FIGURE 2
Yellowness Index Values for Stabilizer Combinations: T₁ and Organotin-borate



38. Figure 2 depicts the results in Table 2. The two bars at the far left of Figure 2 represent the lower (more desirable) yellowness index values achieved with combinations of the claimed invention. The claimed combinations show much less discoloration than the combination of T₁ (a claimed organotin compound) with ODMA (a sulfur compound outside the scope of the claimed invention).

39. In contrast, the superiority of the reverse esters as co-stabilizers is not nearly as evident for combinations of the sulfur compounds with the organotin-borate. The three bars at the far right of Figure 2 show uniformly unacceptable levels of discoloration (all above 30). These results would

not have prompted researchers in the PVH stabilization industry to select a reverse ester over other sulfur compounds disclosed in the Gough patent.

40. I compared the stabilization results for (1) combinations of the organotin-borate (from Gough) and reverse ester sulfur compounds with (2) claimed combinations of an organotin-sulfur compound (T_2) and reverse esters. The claimed organotin-sulfur compound has approximately half the tin content of Gough's organotin-borate compound (which is outside the scope of the claims). I also tested these tin compounds with a sulfur compound outside the scope of the claims, ODMA. These results are shown below in Table 3.

TABLE 3
Whiteness Index Values for Stabilizer Combinations: T_2 and Organotin-borate

| Sulfur Compounds | T_2 ($\frac{1}{2}$ tin content) + Sulfur Compound | Organotin-borate + Sulfur Compound |
|----------------------------------|--|------------------------------------|
| 2-mercaptoethyl stearate (MES) | 5.5 | -13.7 |
| 2-mercaptoethyl octanoate (MEO) | 2.1 | -9.5 |
| octadecyl mercaptoacetate (ODMA) | -28.4 | -22.6 |

Note: The *higher* the number, the better the result.

41. Results for the claimed combinations are shown in bold-face type. In Table 3, it is *very surprising* that claimed combinations with T_2 are much better stabilizers than combinations with the organotin-borate compound because T_2 has only about half the tin content (about 15%) of the organotin-borate (about 30%). The claimed combinations (with T_2) produced whiteness index

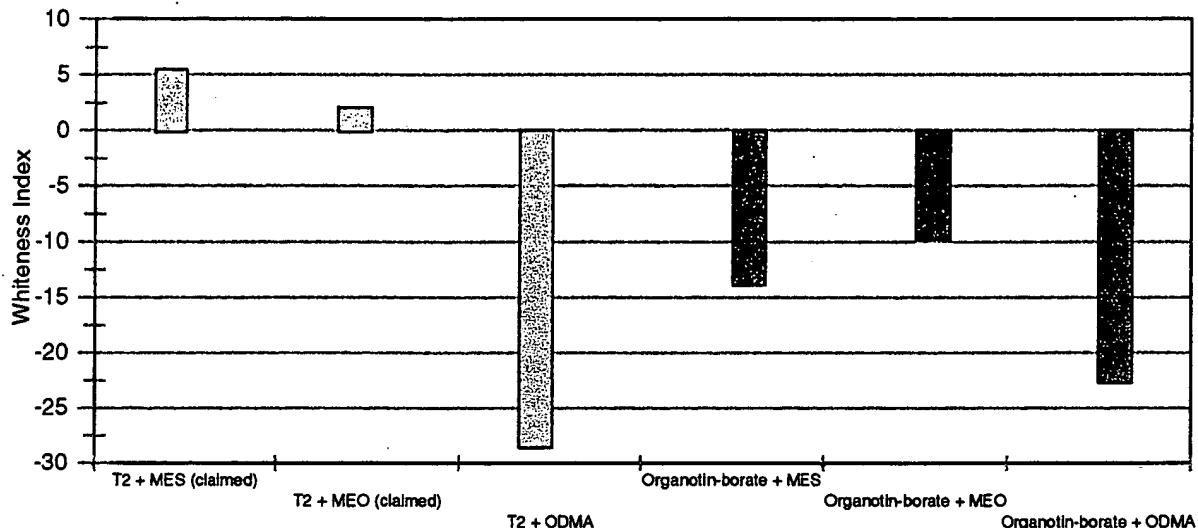
values of 5.5 and 2.1, which are well above the negative whiteness index values of -13.7 and -9.5 for the organotin-borate with the same sulfur compounds.

42. The organotin-borate combinations show uniformly unacceptable whiteness index values, all well below zero. Someone would not have selected reverse esters as particularly promising for stabilizer combinations based on Gough's disclosure or these results.

43. The difference between the values for combinations of T_2 with MES (5.5) and ODMA (-28.4) is extremely surprising. The total difference of 33.9 units is unexpected because the only significant difference between these compounds, as discussed earlier, is the position of the SH group relative to the ester (CO_2) group, a characteristic of reverse esters.

44. The claimed combinations (with reverse esters) give significantly better results than the combination of a claimed tin compound with a sulfur compound outside the scope of the claimed invention, ODMA. The differences between the effects of reverse esters and other sulfur compounds is very striking even when the tin content has been reduced by half, indicating the unique capabilities of the claimed invention to stabilize polymers even with very low levels of a primary tin stabilizer. This ability to reduce tin content translates into significant cost savings.

FIGURE 3
Whiteness Index Values for Stabilizer Combinations: T_2 and Organotin-borate



45. Figure 3 visually depicts the values in Table 3. The claimed combinations, represented by the two bars at the far left of Figure 3, are the only combinations which give whiteness index values above zero. Although these values are still quite low, T_2 only has half the tin content of the organotin-borate and would be expected to be a less effective stabilizer. Instead, the opposite is observed: whereas the claimed combination produces sufficient stabilization effects, the organotin-borate combinations uniformly give unacceptable whiteness index values, whether in combination with reverse esters or another sulfur compound outside the scope of the claimed invention.

46. The significance of the superiority of the reverse esters as co-stabilizers is not observed until the reverse esters are used in combination with a claimed tin stabilizer like T_2 . Clearly, when

combined with T_2 , the two reverse esters at the far left of Figure 3 outperform ODMA, a sulfur compound outside the scope of the claimed invention.

47. Table 4 presents yellowness index results for the same combinations as Table 3.

TABLE 4
Yellowness Index Values for Stabilizer Combinations: T_2 and Organotin-borate

| Sulfur Compounds | T_2 (1/2 tin content) + Sulfur Compound | Organotin-borate + Sulfur Compound |
|----------------------------------|---|------------------------------------|
| 2-mercaptopethyl stearate (MES) | 27.7 | 34.7 |
| 2-mercaptopethyl octanoate (MEO) | 29.1 | 32.6 |
| octadecyl mercaptoacetate (ODMA) | 42.4 | 39.1 |

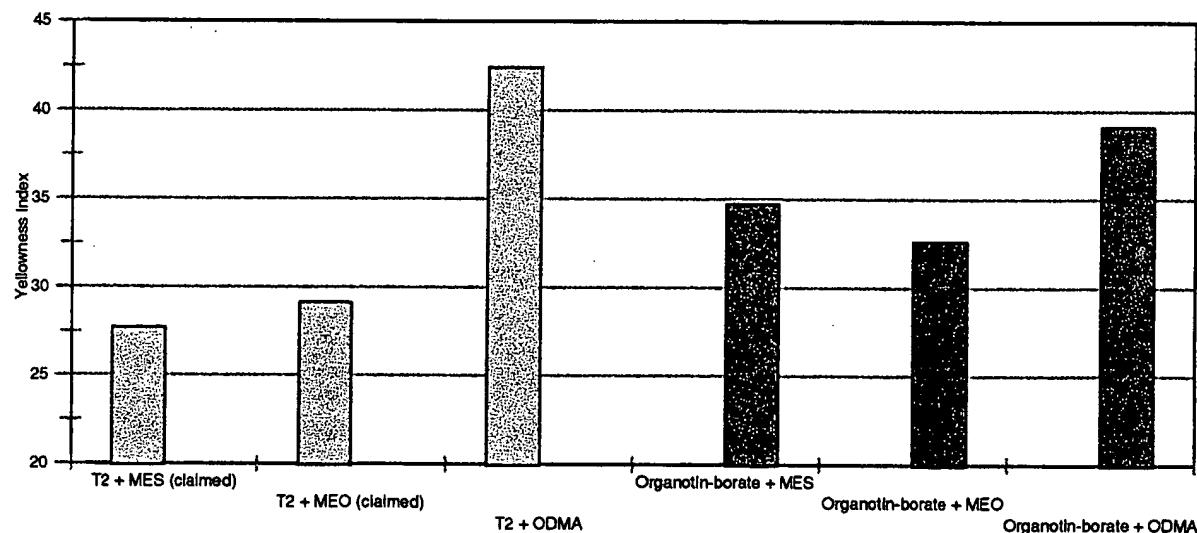
Note: The *lower* the number, the better the result.

48. The yellowness index values in Table 4 are consistent with the whiteness index values in Table 3 for the same combinations of compounds. The comparison between T_2 and the organotin-borate is striking, because T_2 shows somewhat better performance despite T_2 having only about half the tin content (about 15%) of the organotin-borate (about 30%). The claimed combinations, which include T_2 , give yellowness index values of 27.7 and 29.1, show less discoloration than the yellowness index values for combinations of organotin-borate with the claimed sulfur compounds (34.7 and 32.6). Researchers in the PVH stabilization industry would not have expected that such superior yellowness index values could be achieved while reducing the tin content by half.

49. Combinations of the claimed invention are also superior to combinations of T_2 with a sulfur compound outside the scope of the claims, ODMA. The ODMA gives terrible stabilization results

when combined with both the organotin-borate (yellowness index value of 39.1) and T₂ (42.4). Those in the PVH stabilization industry, based on the disclosure of Gough and the similarity of the two compounds, would have expected similar stabilization with ODMA and MES. The only significant difference between these two compounds, as discussed earlier, is the position of the thiol (SH) group relative to the ester (CO₂) group. Surprisingly, with all other reaction reagents and conditions equal, MES gives a value of 27.7, showing significantly less discoloration than ODMA (42.4), a sulfur compound outside the scope of the pending claims.

FIGURE 4
Yellowness Index Values for Stabilizer Combinations: T₂ and Organotin-borate



50. Figure 4 is a graph of the values of Table 4. The yellowness index values of the sulfur compounds in combination with the organotin-borate are all uniformly high (the far right three bars,

all above 30). The values of the claimed invention, the two bars at the far left of Figure 4, are much more acceptable, despite T_2 's significantly lower tin content.

51. The striking difference between the performance of reverse esters and another similar sulfur compound is not evident when the sulfur compounds are tested with organotin-borates. When tested with T_2 , however, the reverse esters are far superior to the sulfur compound outside of the claimed invention, ODMA. The two bars at the far left of Figure 4 show much less discoloration than the third bar from the left of Figure 4 for ODMA.

52. For the Experiments reported in Tables 5-6 and Figures 5-6, the organotin stabilizers were tested at 0.15 phr without the use of a secondary sulfur stabilizer compound. These control experiments demonstrate the ability of the organotin compounds to stabilize PVC alone. Thus, we can observe the relative stabilization or destabilization by the addition of sulfur compounds to these tin compounds in other experiments.

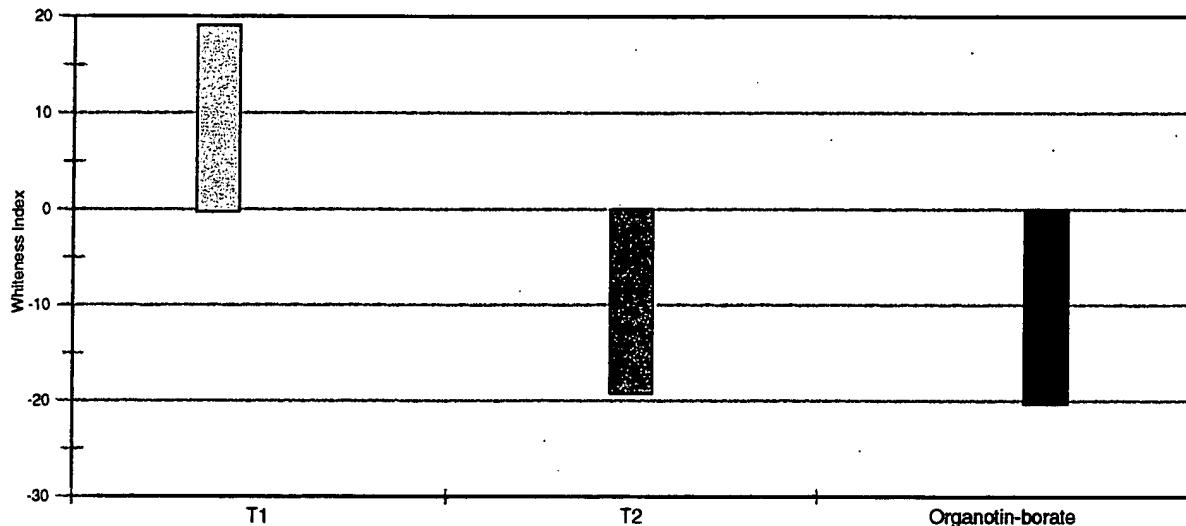
TABLE 5
Whiteness Index Values for Stabilizer Control Experiments

| Controls | T_1 | T_2 ($\frac{1}{2}$ tin content) | Organotin-borate |
|-----------------|-------|------------------------------------|------------------|
| Whiteness Index | 19.1 | -18.9 | -20.1 |

Note: The *higher* the number, the better the result.

53. From Table 5, it is observed that the organotin-borate is a terrible stabilizer by itself. T_1 , with approximately the same tin content as the organotin-borate, is a much better stabilizer alone. In contrast, at the level of tin used in these tests, T_2 by itself gives bad results. However, T_2 has only half the tin content as T_1 or the organotin-borate.

FIGURE 5
Whiteness Index Values for Stabilizer Control Experiments



54. Figure 5 graphically demonstrates whiteness index values from Table 5 for the tin stabilizer compounds used alone. At the levels of tin used in these tests, both the organotin-borate and T_2 are very poor stabilizers alone, although T_2 has about half the tin content and would be expected to be much worse on that basis.

TABLE 6
Yellowness Index Values for Stabilizer Control Experiments

| Controls | T_1 | T_2 ($\frac{1}{2}$ tin content) | Organotin-borate |
|------------------|-------|------------------------------------|------------------|
| Yellowness Index | 23.4 | 41.4 | 59.9 |

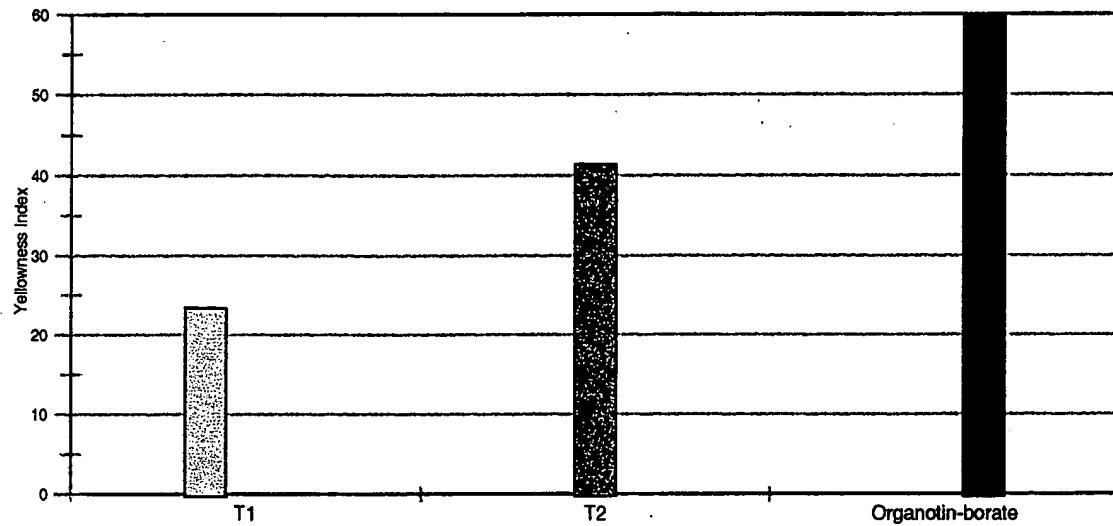
Note: The *lower* the number, the better the result.

55. Table 6 shows even more convincingly than Table 5 what a poor stabilizer the organotin-borate is: the yellowness index value for the organotin-borate, 59.9, is extremely high. This value

is much higher than T_1 (23.4) and it is surprisingly higher (less desirable) than a tin compound in which the tin level is reduced by half (41.4).

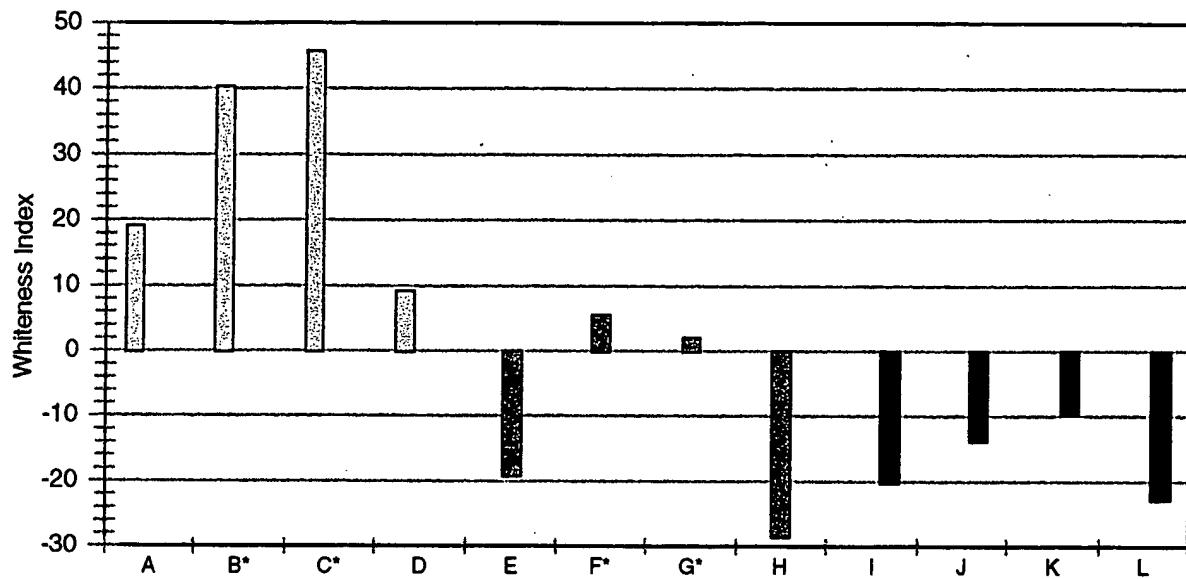
56. These values also show the difficulty of measuring stability in polymers that are very badly degraded. Although the whiteness index value for the organotin-borate (-20.1) was similar to but slightly worse than T_2 (-18.9), the yellowness index value for the borate (59.9) is much worse than the value for T_2 (41.4). As I explained earlier in this Declaration (¶ 22), the colorimeter is not as able to accurately measure the whiteness of a badly degraded polymer as it is able to measure the fine distinctions of whiteness between polymers that are still relatively white. The high yellowness index value, however, is a better indication that the organotin-borate compound is much more discolored and degraded than T_2 , despite T_2 's lower tin content.

FIGURE 6
Yellowness Index Values for Stabilizer Control Experiments



57. Figure 6 graphically depicts the results from Table 6. In my opinion, the organotin-borate is a very poor stabilizer.

FIGURE 7
Whiteness Index Values for Stabilizer Combinations and Tin Control Stabilizers



| | | | | | |
|----|--------------|----|--------------|---|-------------------------|
| A | T_1 | E | T_2 | I | organotin-borate |
| B* | $T_1 + MES$ | F* | $T_2 + MES$ | J | organotin-borate + MES |
| C* | $T_1 + MEO$ | G* | $T_2 + MEO$ | K | organotin-borate + MEO |
| D | $T_1 + ODMA$ | H | $T_2 + ODMA$ | L | organotin-borate + ODMA |

Note: an asterisk (*) indicates combinations within the scope of the claims.

58. Figure 7 depicts all of the preceding data for my twelve experiments. This Figure shows that the claimed reverse esters drastically improve the stabilization of the claimed organotin compounds. It is surprising that the claimed reverse esters (MES and MEO) provide the superior results shown in Figure 7, when compared to the performance of the claimed organotins alone and combined with ODMA.

59. Figure 7 shows the unpredictability of sulfur compounds to either increase or decrease the performance of organotin stabilizers.

60. T_1 and its combinations with sulfur compounds are shown as the four bars at the far left of the figure. The use of T_1 alone is the furthest left. The addition of the claimed reverse esters MES and MEO improves the performance of this compound tremendously. In each case, the whiteness index values increase by over 20 units. The addition of a different type of sulfur compound outside the scope of the claimed invention, ODMA, however, decreases the performance to a level below that of T_1 alone. Such results are typical of the unpredictability of stabilizer combinations. Researchers in the PVH industry do not know if certain classes of compounds will behave synergistically with one another or otherwise except by experimenting.

61. This unpredictability is especially surprising because, as discussed earlier in this Declaration, the only major structural difference between one of the claimed reverse esters, MES, and ODMA is the position of the SH group relative to the CO_2 portion of the molecule, a positional difference which is a characteristic of all the claimed reverse esters. Yet the MES improves the performance of T_1 by 21.2 units, whereas the ODMA actually *decreases* the performance of T_1 by 9.9 units. Nothing in the Gough reference indicates that these different types of compounds would yield such remarkably different results (31.1 total units difference) when used as secondary stabilizers under identical conditions.

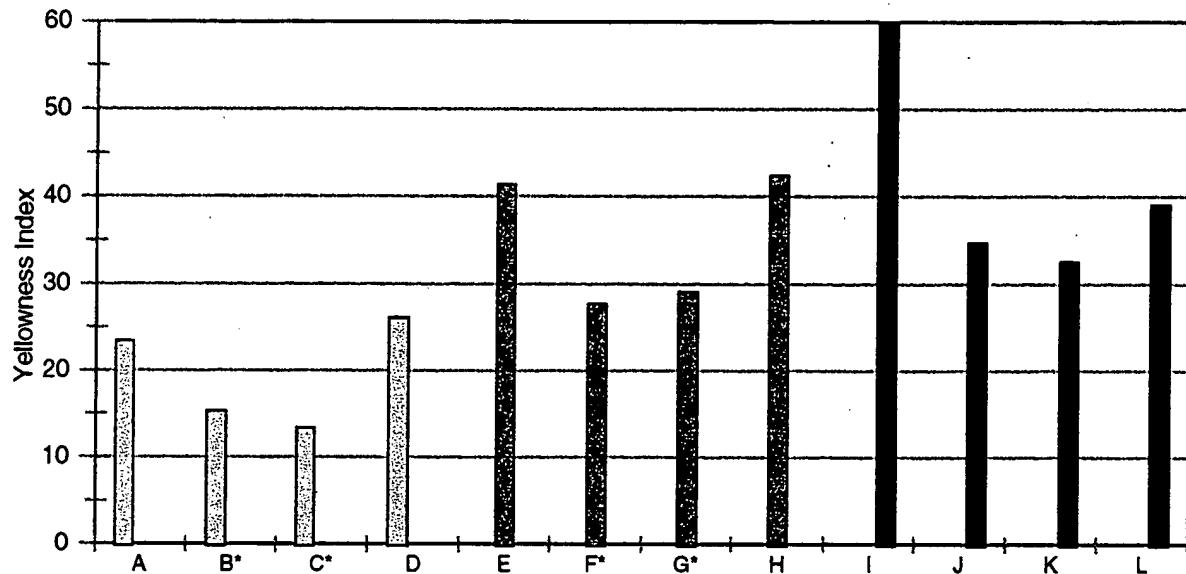
62. Indeed, even if Gough had tested these sulfur compounds, these striking differences between MES and ODMA would not have been observed. As the results for the organotin-borate and its combinations at the far right of Figure 7 show, the combinations with sulfur compounds give

uniformly terrible results (all below zero). All the sulfur compounds would have been dismissed as secondary stabilizers because of their inability to improve Gough's organotin-borate to an acceptable level.

63. Similarly, the addition of sulfur compounds to T_2 yields unexpected results in light of Gough's teaching of the equivalence of sulfur compounds. The results for T_2 are shown as the middle four bars of Figure 7. The absolute values for the T_2 experiments are low, which is not unexpected because T_2 has about half the tin content of T_1 . The tests nevertheless demonstrate the same trends observed in the tests for T_1 .

64. The addition of the two reverse esters improves the performance of T_2 dramatically, in each case increasing the whiteness value of T_2 alone by more than 20 units. The addition of ODMA, a sulfur compound outside the scope of the claims, however, actually decreases the stabilization of T_2 alone. Those in the PVH industry would not have been able to predict on the basis of Gough or any other prior art reference that a reverse ester such as MES would improve the performance of T_2 by 24.4 units, whereas adding ODMA, would decrease the performance of T_2 by 9.5 units. This total difference in performance of 33.9 units is especially surprising because the only significant difference between these two compounds is the fact that one of them is a claimed reverse ester and one of them is not.

FIGURE 8
Yellowness Index Values for Stabilizer Combinations and Tin Control Stabilizers



| | | | | | |
|----|--------------|----|--------------|---|-------------------------|
| A | T_1 | E | T_2 | I | organotin-borate |
| B* | $T_1 + MES$ | F* | $T_2 + MES$ | J | organotin-borate + MES |
| C* | $T_1 + MEO$ | G* | $T_2 + MEO$ | K | organotin-borate + MEO |
| D | $T_1 + ODMA$ | H | $T_2 + ODMA$ | L | organotin-borate + ODMA |

Note: an asterisk (*) indicates combinations within the scope of the claims.

65. Figure 8 graphically depicts the yellowness index values for each of the twelve experiments I performed.

66. Figure 8 confirms my conclusions from Figure 7 about how difficult it is to predict stabilization performance between combinations of sulfur compounds and conventional tin stabilizers like T_1 and T_2 .

67. T_1 and its combinations with sulfur compounds are shown as the 4 bars at the far left of Figure 8. The use of T_1 alone is the furthest left. The addition of the claimed reverse esters, MES and MEO, improves the performance of this compound tremendously. In each case, the yellowness index values are much lower than the control T_1 alone. The addition of ODMA, a sulfur compound outside the scope of the claimed invention, however, increases the yellowness above that obtained from T_1 alone. Experimentation is necessary to determine which combinations of classes of compounds will behave synergistically or otherwise.

68. The unpredictability is even more pronounced because the only major structural difference between two of the sulfur compounds, MES and ODMA, is the position of the SH group relative to the CO_2 portion of the molecule, a positional difference which is a characteristic of all the claimed reverse esters. Yet these compounds with T_1 show a difference in yellowness of (26.1 - 15.3) 10.8 total. Nothing in the Gough reference indicates that these different types of compounds would yield such different results when used as secondary stabilizers under identical conditions.

69. Indeed, as was the case with the whiteness index results, even if Gough had tested these sulfur compounds, these striking differences between MES and ODMA would not have been observed. As the results for the organotin-borate compounds at the far right of Figure 8 show, the addition of sulfur compounds gives uniformly terrible results (all above 30 on the yellowness index). While some improvement is noted with the addition of sulfur compounds, the difference between the reverse esters and ODMA is not remarkable. All the sulfur compounds would have been dismissed as secondary or co-stabilizers because of their inability to improve Gough's organotin-borate to an acceptable level.

70. Similarly, the addition of the sulfur compounds to T_2 yields unexpected results in light of Gough's general teaching of the equivalence of sulfur compounds. The results for T_2 are shown as the middle four bars of Figure 8. The addition of the two reverse esters improves the performance of T_2 dramatically, in each case decreasing the yellowness value by more than 10 units. In contrast, the addition of the sulfur compound ODMA (outside the scope of the claims) to T_2 actually increases the yellow discoloration of T_2 alone.

71. Those in the PVH stabilization industry would not have been able to predict on the basis of Gough or any other prior art reference that a reverse ester such as MES would improve the performance of T_2 by $(41.1 - 27.7)$ **13.7** units, whereas adding ODMA would decrease the performance of T_2 by $(41.4 - 42.4)$ **1** unit. This total difference in performance of **14.7** units is especially surprising because the only significant difference between these two compounds is the fact that one of them is a claimed reverse ester and one of them is not.

IV. Conclusions

72. In my opinion, the claimed combinations are significantly superior to combinations of organotin-borates with reverse esters. The claimed combinations are even superior to combinations of organotin-borates with reverse esters when the tin content of the claimed combination is half that of the organotin-borate combination.

73. Further, it is my opinion that the claimed combinations of the organotin stabilizers with reverse esters are significantly and surprisingly superior to other combinations of tin stabilizers with sulfur compounds that are not reverse esters. Nothing in the cited prior art suggested this remarkable activity of reverse esters relative to other sulfur compounds outside the scope of the claimed invention. Nothing in the prior art would have motivated the selection of combinations of reverse esters with the organotin-sulfur or organotin-halogen compounds of the claimed invention.

I declare under penalty of perjury that the foregoing is true and correct.

By:

Christopher A. Bertolo

Date: May 2, 2000